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Practitioner's Docket No.: 791_130 RCE

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of: Li YANG and Toshihiro YOSHIDA

Ser. No.: 09/770,725

Group Art Unit: 1746

Filed: January 26, 2001

Examiner: Jonathan Crepeau

Confirmation No.: 6015

For: LITHIUM SECONDARY BATTERY

Mail Stop Appeal Brief-Patents
Commissioner for Patents
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Janet M. Stevens
Janet M. Stevens

TRANSMITTAL OF REPLY BRIEF

Sir:

Transmitted herewith are three copies of a Reply Brief for the above-referenced application.

The Commissioner is hereby authorized to charge any additional fees associated with this communication or credit any overpayment to Deposit Account No. 50-1446.

Respectfully submitted,

BURR & BROWN

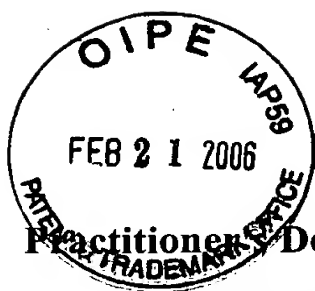
Kevin C. Brown
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Reg. No. 32,402

February 21, 2006
Date

KCB:jms
Enclosures: Reply Brief (3)

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Janet M. Stevens
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REPLY BRIEF

Sir:

The following remarks are in response to the Examiner's Answer dated January 19, 2006.

The Prior Art References Cited in The Statement of The Rejection in The Examiner's Answer Differ From The Prior Art References Listed in The Statement of The Rejection Contained in The Final Office Action

The January 14, 2005 Final Office Action, from which the present Appeal has been taken, included a single rejection which stated "Claims 1-17 are rejected under 35 U.S.C. §103(a) as being unpatentable over Takami et al. (U.S. Patent 6,350,544) in view of Watanabe et al. (U.S. Patent 6,083,644)" (January 14, 2005 Final Office Action, page 2, lines 6-7). The statement of the sole rejection contained in the Examiner's Answer reads "claims 1-17 are rejected under 35 U.S.C. §103(a) as being unpatentable over Takami et al. (U.S. Patent 6,350,544) in view of Watanabe et al. (U.S. Patent 6,083,644) *in view of Kurose et al. (U.S. Patent 6,631,822) as evidence*" [emphasis added](Examiner's Answer, page 3, lines 6-8).

Throughout the prosecution of the present application, it has been unclear as to whether the U.S. PTO is applying Kurose '822 in this rejection or not applying it. For example, in the Final Office Action dated January 14, 2005, there is no mention of Kurose '822 in the statement

of the rejection or in the six paragraphs following the statement of the rejection. The only reference to Kurose '822 in the January 14, 2005 Final Office Action was in a paragraph entitled "Response to Arguments" (January 14, 2005 Office Action, page 3, last four lines and page 4, lines 1-10). It is presumed that the insertion of Kurose '822 into the rejection in the Examiner's Answer was made as a result of recognition that Takami '544 and Watanabe '644, taken individually or in combination, would not have rendered obvious the present invention. The combination of Takami '544, Watanabe '644 and Kurose '822, however, as discussed below, likewise would not have rendered obvious the present invention.

Takami '544, Watanabe '644 and Kurose '822 Taken Individually or in Any Combination Would Not Have Rendered Obvious the Present Invention

The present invention is directed to a lithium secondary battery comprising an electrode unit produced by winding or laminating a positive electrode and a negative electrode via a separator, and a non-aqueous electrolytic solution. The positive electrode comprises active substance comprising lithium manganese oxide. The negative electrode comprises active substance comprising amorphous carbonaceous material and/or graphitized carbonaceous material.

According to the present invention, a cumulative concentration of water released from the positive and negative electrodes in relation to the weight of the electrode unit, exclusive of the weight of the current collectors, is 5,000 ppm or lower in the case of heating both electrodes at 25 to 200°C, and 1,500 ppm or lower in the case of heating the electrodes at 200 to 300°C.

Takami '544 is directed to a lithium secondary battery having, according to the patent, an improved negative electrode containing a carbonaceous material (Takami '544, column 1, lines 9-12). The carbonaceous material has a region of amorphous carbon structure and a region of graphite structure, and has specific properties relating to density and X-ray diffraction. The positive electrode active material of Takami '544 can be any of various kinds of oxides, and a listing of types of oxides appears in Takami '544, column 4, lines 39-49.

Watanabe '644 discloses a non-aqueous electrolyte secondary battery which uses Li_xSiO_y as the negative electrode active material and $\text{Li}_x\text{Ti}_y\text{O}_4$ or Li_xFeS_y as the positive electrode active material (Watanabe '644, Abstract). The materials disclosed in Watanabe '644 for use as the positive and negative electrodes do not include any materials disclosed in Takami '544 for use as

the positive and negative electrodes of Takami '544.

Watanabe '644 contains no suggestion that any of the features disclosed therein which relate to avoidance of moisture would provide any benefit in batteries which do not employ positive and negative active materials as disclosed in Watanabe '644. Watanabe '644 discloses that it is desirable that "*the battery of the present invention*" be assembled in a moisture-free atmosphere or an inert gas atmosphere. Any assertion that Watanabe '644 suggests reducing moisture content in *all* lithium secondary batteries, regardless of the electrode active materials, would clearly go beyond what is disclosed in Watanabe '644.

The Examiner's Answer acknowledges that Takami '544 does not disclose water content of the electrodes as recited in claims 1 and 17 (Examiner's Answer, page 3, lines 3-4 from last). The Examiner's Answer contains a statement that ". . . in column 14, lines 48-52, Watanabe et al. teach that a positive electrode mixture and a negative electrode mixture both have moisture contents of 50 ppm or less" (Examiner's Answer, page 4, lines 4-5). Immediately following the statement quoted in the preceding sentence, the Examiner's Answer contains the statement "[t]herefore, the invention as a whole would have been obvious . . . because the disclosure of Watanabe et al. would motivate the artisan to use electrodes having a moisture content of less than 50 ppm in the battery of Takami et al." (Examiner's Answer, page 4, lines 6-9). Clearly, however, the U.S. PTO has not provided any basis for asserting that one of skill in the art would have been motivated to attempt to modify the batteries disclosed in Takami '544 by looking to Watanabe '644 in the manner asserted in the Examiner's Answer. Moreover, the respective references, Takami '544 and Watanabe '644, do not contain disclosure which would have provided such motivation.

As noted above, the Examiner's Answer refers to U.S Patent No. 6,361,822 (Kurose '822). Kurose '822 is directed to a method for producing an electrode for a non-aqueous electrolyte battery, and more particularly, to a method for producing a non-aqueous electrolyte battery electrode which includes an electrode active material layer containing at least an active material having a composition of $\text{Li}_x \text{Ni}_y \text{M}_z \text{O}_2$ (where x satisfies $0.8 < x < 1.5$, $y+z$ satisfies $0.8 < y+z < 1.2$, z satisfies $0 \leq z < 0.35$, and M is at least one element selected from Co, Mg, Ca, Sr, Al, Mn and Fe).

Kurose '822 says nothing about batteries as described in Takami '544. Kurose '822 does not contain disclosure which would suggest to one of skill in the art that disclosure in Watanabe '644 about the electrodes therein would be broadly applicable to *all* electrodes. As noted above,

Watanabe '644 discloses a battery in which a negative electrode active has the compositional formula Li_xSiO_y , and a positive electrode active material is represented by the general formula $\text{Li}_x\text{Ti}_y\text{O}_4$ or Li_xFeS_y .

The Examiner's Answer contains a statement that "Kurose specifically identifies 'nickel-containing lithium composite oxides' as 'tend[ing] to absorb moisture'" and the Examiner's Answer includes a statement that nickel-containing lithium composite oxides "can be considered to be substantially equivalent to LiMn_2O_4 ."

The Applicants respectfully disagree with the U.S. PTO. As demonstrated by the attached Declaration Under 37 C.F.R. §1.132 (which was entered in this application on July 22, 2005), nickel-containing lithium composite oxides do not behave in a manner anywhere near equivalent to LiMn_2O_4 . In this Declaration, in Experiment 1, electrolyte solution was heated in order to analyze the amount of HF contained in the respective electrolyte solutions after heating. In Experiment 2, respective positive electrode materials (LiNiO_2 and LiMn_2O_4) were placed in respective electrolyte solutions, and the electrolyte solutions were then heated as in Experiment 1, after which the amount of transition metal in the electrolyte solution was detected. Experiment 1 demonstrates that HF is generated by heating the electrolyte solution, similar to the way in which HF is generated by addition of water into the electrolyte solution. HF is believed to enhance the dissolution of transition metals. Table 2 shows that the rate of dissolution of Mn from LiMn_2O_4 is much higher than the rate of dissolution of Ni from LiNiO_2 , indicating that the amount of dissolution of transition metal into the electrolyte solution strongly depends on the type of positive electrode material (i.e., whether it is LiNiO_2 or LiMn_2O_4).

* * * * *

In view of the above, and the Brief on Appeal filed on November 14, 2005, the Honorable Board of Patent Appeals and Interferences is respectfully requested to reverse the final rejection of claims 1-17, and to pass this application to allowance and issuance.

The Commissioner is hereby authorized to charge any additional fees associated with this communication or credit any overpayment to Deposit Account No. 50-1446.

Respectfully submitted,
BURR & BROWN



Kevin C. Brown
Reg. No. 32,402

February 21, 2006
Date

KCB:jms

Attachment:

Declaration Under 37 C.F.R. 1.132 (entered on July 22, 2005)

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Practitioner's Pocket No.: 791_130 RCE

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Serial No.: 09/770,725

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Mail Stop Amendment
Commissioner for Patents
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Alexandria, VA 22313-1450

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Box 1450, Alexandria, VA 22313-1450.

Janet M. Stevens
Janet M. Stevens

DECLARATION UNDER 37 CFR §1.132

Sir:

I, Toshihiro Yoshida, a citizen of Japan hereby declare and state:

1. I have a bachelor's degree in science which was conferred upon me by Tokyo University of Science in Tokyo, Japan, in 1988.
2. I have been employed by NGK INSULATORS, LTD. since 1988 and I have had a total of 7 years of work and research experience in the field of batteries, including lithium secondary batteries.
3. I am one of the inventors in the above-identified patent application and I am familiar with the references applied in the Office Action mailed January 14, 2005.

4. My publications include the following works in this field: "A model of capacity fade caused by a reaction rate between electrolyte and intercalated Li in carbon in Li-ion battery", Proceeding of The 43rd Battery Symposium in Japan (2002) and "A degradation mechanism and life prediction of Li-ion batteries", Proceeding of The 45th Battery Symposium in Japan (2004).

5. The following tests were conducted by me or under my direct supervision.

REPORT OF EXPERIMENT

Purpose of Experiment

The purpose of this experiment is to show the effect of HF in electrolyte solutions upon dissolution of transition-metal contained in lithium transition metal compound oxide, and to clarify the difference in the respective amounts of transition metal dissolved into electrolyte solution between lithium nickel oxide and lithium manganese oxide.

Experimental

Experiment 1: amount of HF in electrolyte solution

Electrolyte solutions (20 ml) each consisting of 1 mol/L $\text{LiPF}_6/\text{EC}+\text{DEC}$ (1:1 in volume) were heated up to 60°C or 100°C for 400 hours. The respective amounts of HF

in the electrolyte solutions thus heated were detected by a titration method, and are shown in Table 1.

Experiment 2: amount of transition metal dissolved

Powders (5g) of positive electrode material were soaked into the electrolyte solution (20 ml), the compositions of which are described above. Two types of the material were used: lithium nickel oxide (LiNiO_2) and lithium manganese oxide (LiMn_2O_4). The electrolyte solutions with powders were heated up to 60°C or 100°C for 400 hours. After filtration of the powders, the amount of transition metal in the electrolyte solution was detected by the ICP method, and are shown in Table 2.

Results and Discussion

Table 1: Amount of HF in Electrolyte Solution Heated

	Amount of HF (PPM)
60°C x 400 hours	313
100°C x 400 hours	2,474

Table 2: Amount of Transition Metal in Electrolyte Solution Heated

	LiNiO_2 Amount of Ni (PPM)	LiMn_2O_4 Amount of Mn (PPM)
60°C x 400 hours	3	22
100°C x 400 hours	4	5,600

Table 1 shows that HF is generated by heating of the electrolyte solution. This phenomenon is similar to that in which HF is generated by the addition of water into an electrolyte solution.

Table 2 shows that the transition metal constituting positive electrode materials dissolves into the electrolyte solution by heating. HF is considered to enhance the dissolution of transition metals. The dissolution rate of Mn is much higher than that of Ni. The difference indicates that the amount of dissolution of transition metal into the electrolyte solution strongly depends on the respective type of positive electrode material.

6. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon

Date: 29 March, 2005

By: T. Yoshida